Supplementary Material. Analytical Solution for Transient Partitioning and Reaction of a Condensing Vapor Species in a Droplet

S.1 Partial Differential Equation for Non-Reactive Species and Its Solution Using Separation of Variables

We begin by considering the transient behavior of a non-reactive gas-phase species that diffuses to the surface of a spherical particle and dissolves in the particle within which it diffuses. The concentration of the species far from the particle is specified. The gas-phase concentration profile around the particle is at steady state. The rate of transport into the particle is governed by the difference between the concentration just above the particle surface and that just inside the particle. Let $C(r,t)$ be the particle-phase concentration of the species of interest, assumed for the moment to be non-reactive, where r is the spatial variable in the radial direction and t is time. Further, D_a is the diffusivity of the species in the particle phase, and A_0 is the concentration at the particle surface required to maintain equilibrium with the atmospheric species concentration at $r = \infty$. The partial differential equation that governs the transient diffusion of the species in the particle is

$$\frac{\partial C(r,t)}{\partial t} = D_a \frac{\partial^2 C(r,t)}{\partial r^2} + \frac{2D_a}{r}\frac{\partial C(r,t)}{\partial r}$$

The boundary and initial conditions are

$$\left[\frac{\partial C(r,t)}{\partial r}\right]_{r=0} = 0\,, \qquad -D_a\left[\frac{\partial C(r,t)}{\partial r}\right]_{r=a} = \gamma[C(a,t) - A_0]\,, \qquad C(r,0) = 0$$

The first boundary condition specifies symmetry around the center of the particle, and the initial condition specifies that at time zero, the particle is free of solute. The second boundary condition expresses the rate of transport of the species into the particle occurs as a result of the difference between the concentration in the gas phase and the equilibrium concentration in the particle phase.

The solution to this problem is given by Crank (1956), as equation (6.40). The steps to obtain this solution are not shown. Because the solution is somewhat involved, we present here the detailed step-by-step solution using separation of variables. To solve this PDE by separation of variables, we first introduce a non-dimensional concentration variable $\Psi(r,t)$ as well as $R(r)$ and $T(t)$, as follows,

$$\Psi(r,t) = \frac{C(r,t) - A_0}{0 - A_0} = R(r)T(t)$$

With this change of variables, the PDE now becomes

$$R(r)\frac{dT(t)}{dt} = D_a T(t)\frac{d^2R(r)}{dr^2} + \frac{2D_a}{r}T(t)\frac{dR(r)}{dr}$$

Dividing through by $R(r)T(t)$ gives

$$\frac{1}{T(t)}\frac{dT(t)}{dt} = D_a\frac{1}{R(r)}\frac{d^2R(r)}{dr^2} + \frac{2D_a}{r}\frac{1}{R(r)}\frac{dR(r)}{dr} = \lambda$$

Since the left hand side of the equation is only a function of t while the right hand side is only a function of r, both sides must be equal to a constant which we call λ. To get the sign of λ, further notice if $\lambda = 0$, then the concentration is constant in time (this is obviously not the case); if $\lambda > 0$, solving the time part of the PDE gives positive exponential dependence of the concentration with respect to time (this system is physically unrealistic and is not considered here). Therefore we conclude that the only condition that gives rise to a physically realistic solution is when $\lambda < 0$.

Now we consider the space component of the two ordinary differential equations. Upon introducing another dimensionless variable ξ and some rearrangement, we have

$$\frac{d^2R(\xi)}{d\xi^2}+\frac{2}{\xi}\frac{dR(\xi)}{d\xi}-\lambda\frac{a^2}{D_a}R(\xi)=0, \qquad \xi=\frac{r}{a}$$

Since λ is negative, the above ODE can be written as

$$\frac{d^2R(\xi)}{d\xi^2}+\frac{2}{\xi}\frac{dR(\xi)}{d\xi}+\beta_n^2R(\xi)=0, \qquad \lambda\frac{a^2}{D_a}=-\beta_n^2, \qquad \beta_n>0$$

To solve this ODE we make another change of variable $\omega(\xi)=R(\xi)\xi$, in which case,

$$\frac{d^2\omega(\xi)}{d\xi^2}=\xi\frac{d^2R(\xi)}{d\xi^2}+2\frac{dR(\xi)}{d\xi}$$

It follows that

$$\frac{d^2R(\xi)}{d\xi^2}+\frac{2}{\xi}\frac{dR(\xi)}{d\xi}=\frac{1}{\xi}\frac{d^2\omega(\xi)}{d\xi^2}=-\beta_n^2R(\xi)$$

which can be rearranged as

$$\frac{d^2\omega(\xi)}{d\xi^2}=-\beta_n^2R(\xi)\xi=-\beta_n^2\omega(\xi)$$

This ODE has the general solution

$$\omega(\xi)=\sigma_1\sin(\beta_n\xi)+\sigma_2\cos(\beta_n\xi),$$

To determine σ_1 or σ_2, recall that B.C. 1 states

$$\left[\frac{\partial C(r,t)}{\partial r}\right]_{r=0}=0$$

In separated and dimensionless form, this translates to

$$\left[\frac{\partial R(\xi)}{\partial \xi}\right]_{\xi=0}=0$$

To determine how B.C. 1 translates to the new variable $\omega(\xi)$, let us first write

$$\frac{d\omega(\xi)}{d\xi}=\xi\frac{dR(\xi)}{d\xi}+R(\xi)$$

At $\xi=0$, $R(\xi)$ is finite, therefore

$$\left[\frac{\partial \omega(\xi)}{\partial \xi}\right]_{\xi=0}$$

is finite. With this bridging result, let us now consider

$$\xi\frac{d\omega(\xi)}{d\xi}=\xi^2\frac{dR(\xi)}{d\xi}+\xi R(\xi)$$

It follows at $\xi=0$, $\omega(\xi)=R(\xi)\xi=0$. And this is the transformed B.C. 1. Using this result, looking back at the general solution of $\omega(\xi)$, we note that $\sigma_2=0$. This allows us to conclude that Φ_R, the eigenfunction of $R(r)$, is

$$\Phi_R=\frac{1}{\xi}\sin\left(\beta_n\xi\right)$$

To determine the restriction on the values β_n, we turn to B.C. 2,

$$-D_a\left[\frac{\partial C(r,t)}{\partial r}\right]_{r=a} = \gamma[C(r=a,t)-A_0]$$

In dimensionless form, this is

$$-D_a\left[\frac{\partial \Psi(r,t)}{\partial r}\right]_{r=a} = \gamma\Psi(a,t)$$

In spatial dimension only, this becomes

$$-D_a\left[\frac{\partial \mathrm{R}(r)}{\partial r}\right]_{r=a} = \gamma\mathrm{R}(a)$$

Recall we concluded that $R(r)$ has the eigenfunction,

$$\Phi_R = \frac{1}{\xi}\sin\left(\beta_n\xi\right) = \frac{a}{r}\sin\left(\beta_n\frac{r}{a}\right)$$

Substituting this result into the equation above, we get

$$\frac{-D_a\beta_n\cos\left(\beta_n\right)}{a} + \frac{D_a\sin\left(\beta_n\right)}{a} = \gamma\sin\left(\beta_n\right)$$

Rearranging, we get the equation that governs the values β_n,

$$\beta_n\cot(\beta_n) + L - 1 = 0, \qquad L = \frac{a\gamma}{D_a}$$

This concludes the space dimension solution of the PDE; to determine out the time dimension, let us return to the time ODE resulted from the separation of variables,

$$\frac{1}{T(t)}\frac{dT(t)}{dt} = \lambda = -\beta_n^2\frac{D_a}{a^2}$$

This equation is easily solved,

$$T(t) = \exp\left(-\beta_n^2\frac{D_a}{a^2}t\right)$$

Thus, the solution of the original PDE is in the form,

$$\Psi(r,t) = \sum_{n=1}^{\infty} A_n\exp\left(-\beta_n^2\frac{D_a}{a^2}t\right)\frac{a}{r}\sin\left(\beta_n\frac{r}{a}\right)$$

To determine the constant coefficient A_n, we need to use the initial condition as well as the orthogonality conditions. At $t = 0$, $\exp(-\beta_n^2 Dt/a^2) = 1$; from the initial condition $C(r,0) = 0$, which in dimensionless form is $\Psi(r,0) = 1$, we have the following relation,

$$1 = \sum_{n=1}^{\infty} A_n\frac{a}{r}\sin\left(\beta_n\frac{r}{a}\right)$$

The orthogonality condition is not entirely obvious, but can be derived from first principles. If we return to the space dimension ODE we derived from separation of variables,

$$\frac{d^2R_n(\xi)}{d\xi^2}+\frac{2}{\xi}\frac{dR_n(\xi)}{d\xi}+\beta_n^2R_n(\xi)=0$$

After some rearrangement, the above expression is equivalent to $(R_n(\xi)\xi)''+\beta_n^2R_n(\xi)\xi=0$. If we now multiply both sides of the equation by $R_m(\xi)\xi$ and integrate from 0 to 1, we have

$$\int_0^1[(R_n(\xi)\xi)''\,(R_m(\xi)\xi)]\,d\xi+\beta_n^2\int_0^1[R_n(\xi)R_m(\xi)\xi^2]\,d\xi=0$$

Very similarly, if we start from index *m*, we get

$$\int_0^1[(R_m(\xi)\xi)''\,(R_n(\xi)\xi)]\,d\xi+\beta_m^2\int_0^1[R_m(\xi)R_n(\xi)\xi^2]\,d\xi=0$$

Using integration by parts,

$$\int_0^1[(R_n(\xi)\xi)''\,(R_m(\xi)\xi)]\,d\xi=\left[\xi R_m(\xi)\{\xi R_n(\xi)\}'\right]_{\xi=0}^{\xi=1}-\int_0^1[(R_n(\xi)\xi)'\,(R_m(\xi)\xi)']\,d\xi$$

$$\int_0^1[(R_m(\xi)\xi)''\,(R_n(\xi)\xi)]\,d\xi=\left[\xi R_n(\xi)\{\xi R_m(\xi)\}'\right]_{\xi=0}^{\xi=1}-\int_0^1[(R_m(\xi)\xi)'\,(R_n(\xi)\xi)']\,d\xi$$

Since $R_n(\xi)$ and $R_m(\xi)$ are the same at the two boundaries $\xi=0$ and $\xi=1$, we conclude that

$$\int_0^1[(R_n(\xi)\xi)''\,(R_m(\xi)\xi)]\,d\xi=\int_0^1[(R_m(\xi)\xi)''\,(R_n(\xi)\xi)]\,d\xi$$

But since $\beta_n^2\neq\beta_m^2$, the orthogonality condition is, unless *n* = *m*,

$$\int_0^1[R_n(\xi)R_m(\xi)\xi^2]d\xi=0$$

Therefore, in order to use this orthogonality condition, we multiply both sides of

$$1=\sum_{n=1}^{\infty}A_n\frac{1}{\xi}\sin(\beta_n\xi)$$

by $\xi^2 * 1/\xi\sin(\beta_m\xi)$ and integrate from 0 to 1

$$\int_0^1\left[\xi^2\frac{1}{\xi}\sin(\beta_m\xi)\right]d\xi=A_n\int_0^1\left[\sum_{n=1}^{\infty}\frac{1}{\xi}\sin(\beta_n\xi)\xi^2\frac{1}{\xi}\sin(\beta_m\xi)\right]d\xi$$

Now by the orthogonality condition,

$$\int_0^1[\xi\sin(\beta_n\xi)]d\xi=A_n\int_0^1[\sin(\beta_n\xi)^2]d\xi$$

Therefore, we have

$$A_n = \frac{4[\beta_n \cos(\beta_n) - \sin(\beta_n)]}{\beta_n[\sin(2\beta_n) - 2\beta_n]} = \frac{2L}{\beta_n^2 + L(L-1)\sin(\beta_n)}, \qquad \text{using } \beta_n \cot(\beta_n) + L - 1 = 0, \qquad L = \frac{a\gamma}{D_a}$$

Assembling everything, the solution of the original PDE is

$$\Psi(r,t) = \frac{C(r,t) - A_0}{0 - A_0} = \sum_{n=1}^{\infty} \frac{2L}{\beta_n^2 + L(L-1)\sin(\beta_n)} \exp\left(-\beta_n^2 \frac{D}{a^2} t\right) \frac{a}{r} \sin\left(\beta_n \frac{r}{a}\right)$$

$$\beta_n \cot(\beta_n) + L - 1 = 0, \qquad L = \frac{a\gamma}{D_a}$$

This result confirms that given by Crank.

S.2 Solution of the Original PDE Including First-Order Reaction

We now employ a method due to Dankwerts that allows one to obtain a solution to the problem involving first-order reaction from the solution for the inert species (Crank, 1956). The Danckwerts method states if $C(r,t)$ is the solution to

$$\frac{\partial C(r,t)}{\partial t} = D_a \frac{\partial^2 C(r,t)}{\partial r^2}$$

then the solution $A(r,t)$ for

$$\frac{\partial A(r,t)}{\partial t} = D_a \frac{\partial^2 A(r,t)}{\partial r^2} - kA(r,t)$$

is given by

$$A(r,t) = k \int_0^t C(r,t') \exp(-kt')\, dt' + C(r,t) \exp(-kt)$$

Using this result, we want to show if $C(r,t)$ is the solution to

$$\frac{\partial C(r,t)}{\partial t} = D_a \frac{\partial^2 C(r,t)}{\partial r^2} + \frac{2D_a}{r} \frac{\partial C(r,t)}{\partial r}$$

with the boundary and initial conditions,

$$\left[\frac{\partial C(r,t)}{\partial r}\right]_{r=0} = 0, \qquad -D_a \left[\frac{\partial C(r,t)}{\partial r}\right]_{r=a} = \gamma[C(a,t) - C_0], \qquad C(r,t=0) = 0$$

then

$$A(r,t) = k \int_0^t C(r,t') \exp(-kt')\, dt' + C(r,t) \exp(-kt)$$

is a solution to

$$\frac{\partial A(r,t)}{\partial t} = D_a \frac{\partial^2 A(r,t)}{\partial r^2} + \frac{2D_a}{r} \frac{\partial A(r,t)}{\partial r} - kA(r,t)$$

with the boundary and initial conditions,

$$\left[\frac{\partial A(r,t)}{\partial r}\right]_{r=0} = 0, \qquad -D_a \left[\frac{\partial A(r,t)}{\partial r}\right]_{r=a} = \gamma[A(a,t) - A_0], \qquad A(r,t=0) = 0$$

We will first show that the solution for $A(r,t)$ satisfies the modified PDE, then show that the boundary and initial conditions are satisfied:

$$A(r,t) = k\int_0^t C(r,t')\exp(-kt')\,dt' + C(r,t)\exp(-kt)$$

$$\frac{\partial A(r,t)}{\partial t} = kC(r,t)\exp(-kt) - kC(r,t)\exp(-kt) + \frac{\partial C(r,t)}{\partial t}\exp(-kt) = \frac{\partial C(r,t)}{\partial t}\exp(-kt)$$

$$\frac{\partial A(r,t)}{\partial r} = k\int_0^t \frac{\partial C(r,t')}{\partial r}\exp(-kt')\,dt' + \frac{\partial C(r,t)}{\partial r}\exp\,(-kt)$$

$$\frac{\partial^2 A(r,t)}{\partial r^2} = k\int_0^t \frac{\partial^2 C(r,t')}{\partial r^2}\exp(-kt')\,dt' + \frac{\partial^2 C(r,t)}{\partial r^2}\exp\,(-kt)$$

It follows that

$$\begin{aligned}
&2\frac{D_a}{r}\frac{\partial A(r,t)}{\partial r} + D_a\frac{\partial^2 A(r,t)}{\partial r^2} \\
&\quad = D_a\left\{k\int_0^t \frac{\partial^2 C(r,t')}{\partial r^2}\exp(-kt')\,dt' + \frac{\partial^2 C(r,t)}{\partial r^2}\exp(-kt) + \frac{2}{r}\,k\int_0^t \frac{\partial C(r,t')}{\partial r}\exp(-kt')\,dt' + \frac{\partial C(r,t)}{\partial r}\exp(-kt)\right\} \\
&\quad = D_a\left\{k\int_0^t \left[\frac{\partial^2 C(r,t')}{\partial r^2}\frac{\partial C(r,t')}{\partial r}\frac{2}{r}\frac{\partial C(r,t')}{\partial r}\right]\exp(-kt')\,dt' + \left[\frac{\partial^2 C(r,t)}{\partial r^2}\frac{\partial C(r,t)}{\partial r}\frac{2}{r}\frac{\partial C(r,t)}{\partial r}\right]\exp(-kt)\right\} \\
&\quad = k\int_0^t \frac{\partial C(r,t')}{\partial t}\exp(-kt')\,dt' + \frac{\partial C(r,t)}{\partial t}\exp(-kt) = k\int_0^t \frac{\partial C(r,t')}{\partial t}\exp(-kt')\,dt' + \frac{\partial A(r,t)}{\partial t},
\end{aligned}$$

$$\text{where } \frac{\partial C(r,t)}{\partial t} = D_a\frac{\partial^2 C(r,t)}{\partial r^2} + \frac{2D_a}{r}\frac{\partial C(r,t)}{\partial r} \text{ and } \frac{\partial A(r,t)}{\partial t} = \frac{\partial C(r,t)}{\partial t}\exp(-kt)$$

It becomes clear that, in order to show

$$\frac{\partial A(r,t)}{\partial t} = D_a\frac{\partial^2 A(r,t)}{\partial r^2} + \frac{2D_a}{r}\frac{\partial A(r,t)}{\partial r} - kA(r,t)$$

or in other words,

$$D_a\frac{\partial^2 A(r,t)}{\partial r^2} + \frac{2D_a}{r}\frac{\partial A(r,t)}{\partial r} = \frac{\partial A(r,t)}{\partial t} + kA(r,t)$$

we need to show that

$$k\int_0^t \frac{\partial C(r,t')}{\partial t}\exp(-kt')\,dt' = kA(r,t)$$

To see this, let us write out the assumed expression for $A(r,t)$,

$$\begin{aligned}
A(r,t) &= k\int_0^t C(r,t)\exp(-kt')\,dt' + C(r,t)\exp(-kt) \\
&= kC(r,t)\left(-\frac{1}{k}\right)\exp(-kt) + k\frac{1}{k}\int_0^t \frac{\partial C(r,t')}{\partial t}\exp(-kt')\,dt' + C(r,t)\exp(-k*t) = \int_0^t \frac{\partial C(r,t')}{\partial t}\exp(-kt')\,dt'
\end{aligned}$$

Thus we have shown that

$$k\int_0^t \frac{\partial C(r,t')}{\partial t}\exp(-kt')\,dt' = kA(r,t)$$

This completes the demonstration that the solution for $A(r,t)$ satisfies the modified PDE.

Next we need to demonstrate the equivalence of the boundary conditions. We note that

$$\left[\frac{\partial A(r,t)}{\partial r}\right]_{r=0} = 0 \text{ and } A(r,t=0) = 0 \text{ as long as } \left[\frac{\partial C(r,t)}{\partial r}\right]_{r=0} = 0 \text{ and } C(r,t=0) = 0,$$

$$\text{for } A(r,t) = k\int_0^t C(r,t')\exp(-kt')\,dt' + C(r,t)\exp(-kt)$$

$$\text{and } \frac{\partial A(r,t)}{\partial r} = \ k\int_0^t \frac{\partial C(r,t')}{\partial r}\exp(-kt')\,dt' + \frac{\partial C(r,t)}{\partial r}\exp\,(-kt)$$

Considering B.C. 2,

$$-D_a\left[\frac{\partial C(r,t)}{\partial r}\right]_{r=a} = \gamma[C(a,t) - A_0]$$

$$\begin{aligned}
\left[\frac{\partial A(r,t)}{\partial r}\right]_{r=a} &= k\int_0^t \left[\frac{\partial C(r,t')}{\partial r}\right]_{r=a}\exp(-kt')\,dt' + \left[\frac{\partial C(r,t)}{\partial r}\right]_{r=a}\exp(-kt) \\
&= \frac{\gamma}{D_a}k\int_0^t [A_0 - C(a,t')]\,dt' + \frac{\gamma}{D_a}[A_0 - C(a,t)]\exp(-kt) \\
&= \frac{\gamma}{D_a}k\int_0^t A_0\exp(-kt')\,dt' + \frac{\gamma}{D_a}A_0\exp(-kt) - \frac{\gamma}{D_a}[A(r,t)]_{r=a} \\
&= \frac{\gamma}{D_a}\left\{kA_0\left[\left(-\frac{1}{k}\right)\exp(-kt) - \frac{1}{k}\right] + A_0\exp(-kt)\right\} - \frac{\gamma}{D_a}[A(r,t)]_{r=a} = \frac{\gamma}{D_a}(A_0 - [A(r,t)]_{r=a})
\end{aligned}$$

In the previous section we concluded that

$$\frac{C(r,t) - A_0}{0 - A_0} = \sum_{n=1}^{\infty}\frac{2L}{\beta_n^2 + L(L-1)\sin(\beta_n)}\exp\left(-\beta_n^2\frac{D_a}{a^2}t\right)\frac{a}{r}\sin\left(\beta_n\frac{r}{a}\right)$$

$$\beta_n\cot(\beta_n) + L - 1 = 0, \qquad L = \frac{a\gamma}{D_a}$$

that is

$$C(r,t) = A_0 + \sum_{n=1}^{\infty}\frac{-2LA_0}{\beta_n^2 + L(L-1)\sin(\beta_n)}\exp\left(-\beta_n^2\frac{D_a}{a^2}t\right)\frac{a}{r}\sin\left(\beta_n\frac{r}{a}\right)$$

Using the Danckwerts transformation, we can now write the full solution to the problem involving first-order reaction as,

$$A(r,t) = k\int_0^t \left\{A_0 + \sum_{n=1}^{\infty}\frac{-2LA_0}{\beta_n^2 + L(L-1)\sin(\beta_n)}\exp\left(-\beta_n^2\frac{D_a}{a^2}t'\right)\frac{a}{r}\sin\left(\beta_n\frac{r}{a}\right)\right\}\exp(-kt')\,dt'$$
$$+\left\{A_0 + \sum_{n=1}^{\infty}\frac{-2LA_0}{\beta_n^2 + L(L-1)\sin(\beta_n)}\exp\left(-\beta_n^2\frac{D_a}{a^2}t\right)\frac{a}{r}\sin\left(\beta_n\frac{r}{a}\right)\right\}\exp(-kt)$$

$$\beta_n\cot(\beta_n) + L - 1 = 0, \qquad L = \frac{a\gamma}{D}$$

S.3 Relating the Constants in the Solution to the Actual Physical Constants

In the original problem statement,

$$\frac{\partial A(r,t)}{\partial t} = D_a\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial A(r,t)}{\partial r}\right) - kA(r,t)$$

$$\left[\frac{\partial A(r,t)}{\partial r}\right]_{r=0} = 0\,, \qquad D_a\left[\frac{\partial A(r,t)}{\partial r}\right]_{r=a} + \frac{v_s}{HRT}A(r,t) = v_sG(\infty,t)\,, \qquad A(r,0) = 0$$

Comparing this with the PDE we have solved,

$$\gamma = \frac{v_s}{HRT}\,, \qquad A_0 = HRTG(\infty,t)$$

S.4 Solution when the Gas-Phase Concentration Decreases with Time due to Uptake by and Reaction in the Particle

In the solution above, it was assumed that the gas-phase concentration A_0 is constant. Of interest, is the case in which the concentration of the species in the gas phase decreases with time owing to its consumption in the particle. A balance on the concentration of the species in the gas phase leads to the following ODE,

$$\frac{dG(\infty,t)}{dt} = -4\pi a^2 ND_a\left[\frac{\partial A(r,t)}{\partial r}\right]_{r=a}, \qquad G(\infty,0) = G_0$$

where N is the number concentration of particles in the air. Using the solution we have obtained, this ODE becomes,

$$\frac{dG(\infty,t)}{dt} = -G(\infty,t)\left\{1 - \left[k\int_0^t\left\{1 - \sum_{n=1}^{\infty}\frac{2L}{\beta_n^2 + L(L-1)}\exp\left(-\beta_n^2\frac{D_a}{a^2}t'\right)\right\}\exp(-kt')\,dt' + \left(\left\{1 - \sum_{n=1}^{\infty}\frac{2L}{\beta_n^2 + L(L-1)}\exp\left(-\beta_n^2\frac{D_a}{a^2}t\right)\right\}\exp(-kt)\right)\right]\right\},$$

$$G(\infty,0) = G_0, \quad \beta_n\cot(\beta_n) + L - 1 = 0, \qquad L = \frac{a\gamma}{D_a}, \qquad \gamma = \frac{v_s}{HRT},$$

$$\delta = \frac{4\pi a^2 ND_a}{D_a}$$

based on the fact that

$$\left[\frac{\partial A(r,t)}{\partial r}\right]_{r=a} = -\frac{\gamma}{D_a}[A(a,t) - A_0]$$

Reference

Crank, J., *The Mathematics of Diffusion*, Oxford University Press, Oxford, UK (1956).